RADIUM-CONTENT OF CEYLON THORIANITE*

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(*Received for publication, Sept. 7, 1943*)

ABSTRACT. A pliotron-amplifier has been set up for the study of many interesting nuclear and radio-active phenomena. With this amplifier and ionisation chamber, the radium content of a sample of Ceylon thorianite has been estimated by the well-known γ-ray method of estimation. The amount of radium present approximately amounts to 5.94×10^{-8} gms. per gm. of the mineral.

INTRODUCTION

An accurate estimation of radium present in minute traces, in different minerals has been already known to be of much commercial importance due to a large demand of this element in medical science and in making luminous paints. Besides that, estimation of radio-active elements and the equilibrium amount of their product elements in different minerals and rocks is of much theoretical interest to geophysicists. As only from such data we can calculate the age of the earth and trace the history of gradual evolution of different rocks and mountains.

The age of a rock or mineral is defined as the time elapsed since the beginning of solidification or crystallisation from the molten state. This time can be easily obtained from the experimentally determined ratio :

$$\text{Age} = \frac{\text{Accumulated amount of disintegration product element}}{\text{Rate of production of the product element}}.$$

The product element is either helium which is nothing but the accumulated α-particles emitted by the successive disintegration products, or radiogenic lead which is the end product of the three radio-active series. The amount of helium or lead present per gram of a specimen is determined by chemical analysis and suitable analytical technique. But recent investigations show that in many cases helium-method gives erroneous results due to the leakage of helium from rocks or minerals. Again as the life of parent radio-active elements, such as uranium or thorium, is very long there is negligible change in the amount of parent element since the beginning of rock-formation. Therefore the rate of production of the product element is directly proportional to the amount of uranium or thorium present in the rock or mineral. This amount can be ascertained by chemical analysis of the sample. But as the amount of uranium or thorium present is very small particularly in case of rocks, the chemical

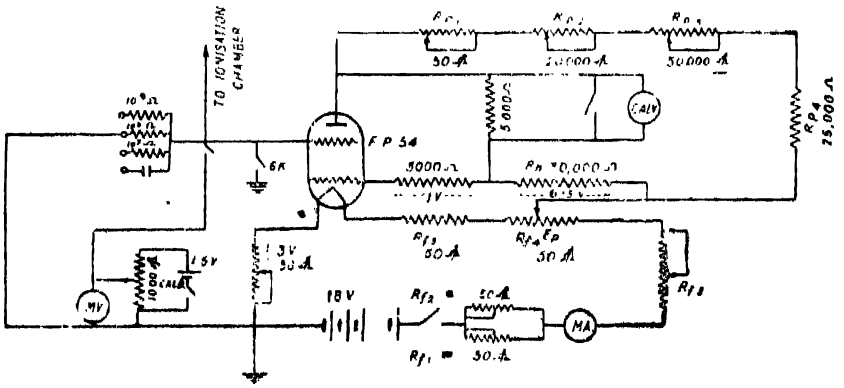
* Communicated by the Indian Physical Society.

method is liable to considerable error. The alternative and much more reliable method developed and followed by Evans (1940), Nier (1941) and all the modern workers is by the comparison of ionisation current produced by certain weight of a specimen with that of a standard source. Usually α- and β-rays are cut off and the ionisation due to γ-ray only is compared. In case of very feeble source, uranium, etc., present can be directly determined by counting α-rays emitted by a thin layer of the sample. The comparison of ionisation current or the counting of α-particles being done by an ionisation chamber with an electrometer or a vacuum-tube amplifier.

The present work was undertaken for a systematic estimation of radium-content and also for the age-determination of Indian minerals and rocks in co-operation with Dr. Fox, Director, Geological Survey of India and Dr. Wadia, Mineralogist to the Government of Ceylon. Both of them very kindly consented to help us in this work.

APPARATUS

The apparatus set up for this experiment is an electrometer-tube amplifier with an ionisation chamber. In recent years such an arrangement has been extensively used for the study of various nuclear and radio-active problems. Due to reliable results and very convenient manipulation it is quickly replacing electrometers and electroscope for the investigation of these problems. The electrometer-tube selected is an F. P. 54, pliotron tube designed by Metcalf and Thompson (1930). The circuit used is same in principle as that of Dubridge and Brown (1933). A Leeds and Northroup R-type of galvanometer was balanced by the plate current and the space-charge grid current of the tube. To eliminate drift and for fine control, various resistances were selected by trial. The actual circuit set up is shown in Fig. 1. Further to eliminate mechanical disturbances the ionisation chamber was carefully mounted on the Pliotron-tube enclosed in a brass cylinder. The mounting is shown in Fig. 2. Using 10^9 ohms as the leaking resistance a sensitivity nearly 10^{-14} amp. per millimeter deflection was obtained.



F. P. 54 - PLIOTRON AMPLIFIER CIRCUIT

FIG. 1

THE ESTIMATION OF RADIUM IN THE MINERAL THORIANITE OF CEYLON

The mineral under investigation was sent by Dr. Wadia on behalf of the Government of Ceylon for the estimation of radium-content in the mineral. A particular sample of the Ceylon Thorianite has been chemically analysed as early as about 1910 by Soddy (1910) and as referred by him, the sample contains 60 to 70% of ThO_2 and from 10 to 20% of U_3O_8. The mineral will therefore contain both radium and its isotope mesothorium 1. From this chemical analysis, the approximate amount of radium and mesothorium 1 present can be theoretically obtained from the consideration of radio-active equilibrium. As it is interesting to compare the theoretical amount with that estimated by physical process, we have calculated the theoretical amount as shown below. When a radio-active element is in equilibrium with its product elements then from the law of radio-active disintegration it follows that

$$(N_1\lambda_1)_{\overline{U}} = (N_2\lambda_2)_{Ra} \quad \ldots \quad (1)$$

$$(N_1\lambda_1)_{Th} = (N_2\lambda_2)_{MsTh1} \quad \ldots \quad (2)$$

where N_1 represents the number of uranium and thorium atoms present in the respective cases and N_2 that of radium and mesothorium 1 atoms present in equilibrium, λ's represent the decay constant of the respective element.

Let us first calculate the radium-content per gm. of the mineral. It is experimentally known that $\lambda_U = 4.9 \times 10^{-18}$ sec^{-1}, $\lambda_{Ra} = 1.373 \times 10^{-11}$ sec^{-1}; and atomic wt. of U = 238 and of Ra = 226. If the percentage of U_3O_8 present be assumed to be 15%, the mean value of the chemical analysis, then 1 gm. of the mineral will contain $\frac{3 \times 714}{20 \times 842}$ gms. of U. Since 238 gms. of uranium contain 6.06×10^{23} atoms (Avogadro number) therefore 1 gm. of the mineral will contain $\frac{3 \times 714 \times 6.06 \times 10^{23}}{20 \times 842 \times 238}$ atoms of U. Therefore if there are N_{Ra} atoms of radium in equilibrium then from (1) it follows that

$$\frac{3 \times 714 \times 6.06 \times 10^{23}}{20 \times 842 \times 238} \times 4.9 \times 10^{-18} = N_{Ra} \times 1.37 \times 10^{-11}$$

$$\therefore \quad N_{Ra} = \frac{3 \times 714 \times 6.06 \times 4.9}{20 \times 842 \times 238 \times 1.37} \times 10^{16} \text{ atoms.}$$

Again 226 gm. of radium contain $(6.06) \times 10^{23}$ atoms therefore the wt. of N_{Ra} atoms of radium is

$$\frac{3 \times 714 \times 6.06 \times 4.9 \times 226 \times 10^{16}}{20 \times 842 \times 238 \times 1.37 \times 6.06 \times 10^{23}} = 4.32 \times 10^{-8} \text{ gms.}$$

$\therefore$ One gm. of the mineral contains about 4.32×10^{-8} gms. of radium.

Similarly if the percentage of ThO_2 is 70, then from the experimental value of $\lambda_{Th} = 1.33 \times 10^{-18}$ sec^{-1}, $\lambda_{MsTh_1} = 3.28 \times 10^{-9}$ sec^{-1} and the atomic wt. of

thorium and mesothorium are 232 and 228 respectively, it follows that 1 gm. of the mineral contain $\frac{228 \times 70 \times 1.33 \times 10^{-11}}{264.4 \times 3.28} = 2.45 \times 10^{-10}$ gms. of MsThI.

From this calculation it follows that although the percentage of ThO_2 is much higher than that of U_3O_8 still the equilibrium amount of mesothorium 1 is much smaller, almost negligible in comparison with that of radium. This is however due to the fact that the half life of MsThI is much shorter than that of radium.

EXPERIMENTAL PROCEDURE

The radium-content of the mineral has been estimated by the well-known γ-ray method of estimation. An ionisation chamber together with the pliotron amplifier described above has been used to compare the ionisation current due to γ-rays of a certain weight of the mineral with that of a standard γ-ray source, under exactly similar conditions. The ionisation chamber constructed is a lead cylinder about 5″ long and of internal diameter about 2″. The thickness of the wall is 1 cm. The collecting electrode is a brass wire of diameter about 1/16″, and insulated from the wall by an amber cork. About 250 volts between the wall and the central electrode was found sufficient to produce saturation current due to ionisation. The collecting electrode was connected with the grid of the pliotron tube and the mounting was done as shown in the Fig. 2. The standard sample used is 0.47 milligram of radium in the form of radium bromide. The ionisation current was compared by comparing the deflection in the galvanometer scale. The approximate linearity of the deflection was tested by taking the standard source at different distances from the ionisation chamber.

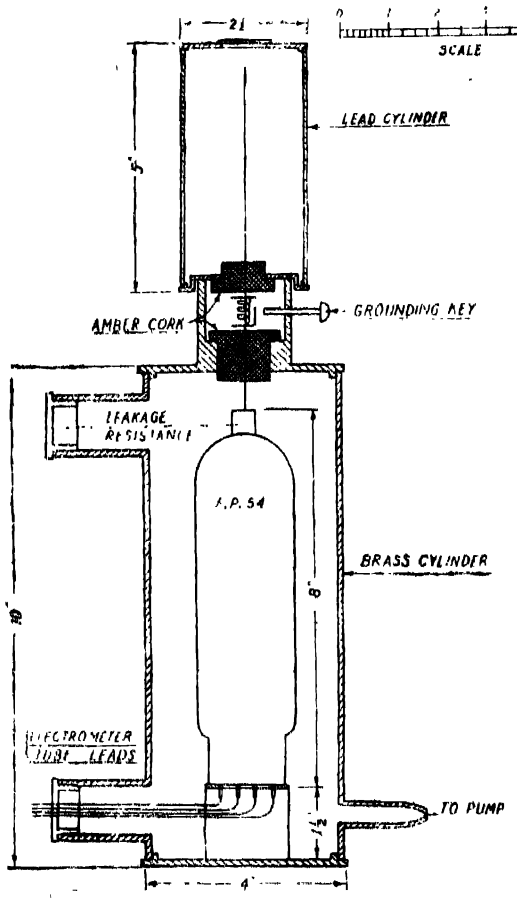


FIG. 2

EXPERIMENTAL DATA

The mean deflection for 0.47 milligram of radium 255 mm.

" " 217 gms. of thorianite 7 mm.

Radium content of per gm of the mineral $= \frac{47 \times 7 \times 10^{-3}}{250 \times 217} = 5.94 \times 10^{-8}$ gm.

As the mineral was spread in a thin layer of about $\frac{1}{2}$ cm. the correction due to self-absorption was small and therefore neglected.

From this experimental estimation it follows that the radium-content per gm. of the mineral of this sample is appreciably greater than that obtained theoretically from Soddy's chemical analysis assuming the percentage of U_3O_8 to be 15%. The percentage of U_3O_8 appears to be 20.63% which is very near to that of the upper limit of Soddy's estimation.

When this work was undertaken it was also the idea to estimate the age of formation of the rock. As already stated the age can be easily calculated from the well-known method of lead-uranium ratio. When radium is estimated by γ-ray method, the amount of uranium present can be calculated by the consideration of radio-active equilibrium. But for the estimation of lead, a chemical analysis of the sample is necessary. Moreover the lead present in the mineral is generally a mixture of different isotopes of lead produced by the decay of uranium, thorium, and actinium and probably some trace of ordinary lead. Therefore a mass-spectrographic analysis of the lead is necessary to ascertain the percentage of uranium or any other type of lead. For all these difficulties the work could not be proceeded further.

In conclusion I express my thanks to Professor M. N. Saha, F.R.S., for his kind guidance to complete this work. My thanks are particularly due to Dr. Wadia, Mineralogist to the Government of Ceylon for kindly supplying the mineral Thorianite and permitting me to publish the radium estimation of the sample with this apparatus. My thanks are also due to Dr. B. D. Nag Choudhury and Dr. S. C. Sirkar for many helpful suggestions. I also acknowledge my thanks to Dr. D. M. Bose, Director, Bose Research Institute, Calcutta, for kindly permitting me to complete the work.

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